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Title: Analytical and Radiochemistry for Nuclear Forensics

Author(s): Steiner, Robert Ernest
Dry, Donald E.
Kinman, William Scott
Podlesak, David
Tandon, Lav

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Analytical and Radiochemistry for Nuclear Forensics

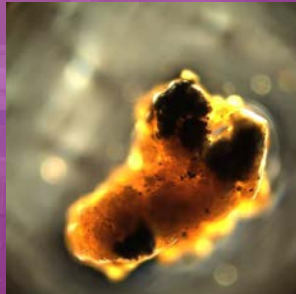
Robert Steiner, Don Dry, William Kinman,
Dave Podlesak and Lav Tandon

DOC-9: RN91-311-002

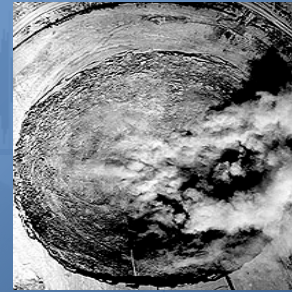
Overview

- Introduction to nonproliferation nuclear forensics
- Scope of activities in forensics at Los Alamos National Laboratory
- Facilities for radioanalytical work at LANL
- Radiochemical characterization capabilities
- Bulk chemical and materials analysis capabilities
- Future interests in forensics interactions

Technical Nuclear Forensics: LANL technical capabilities fostered by the nuclear weapons program support the forensic evaluation of all categories of nuclear events:



Materials

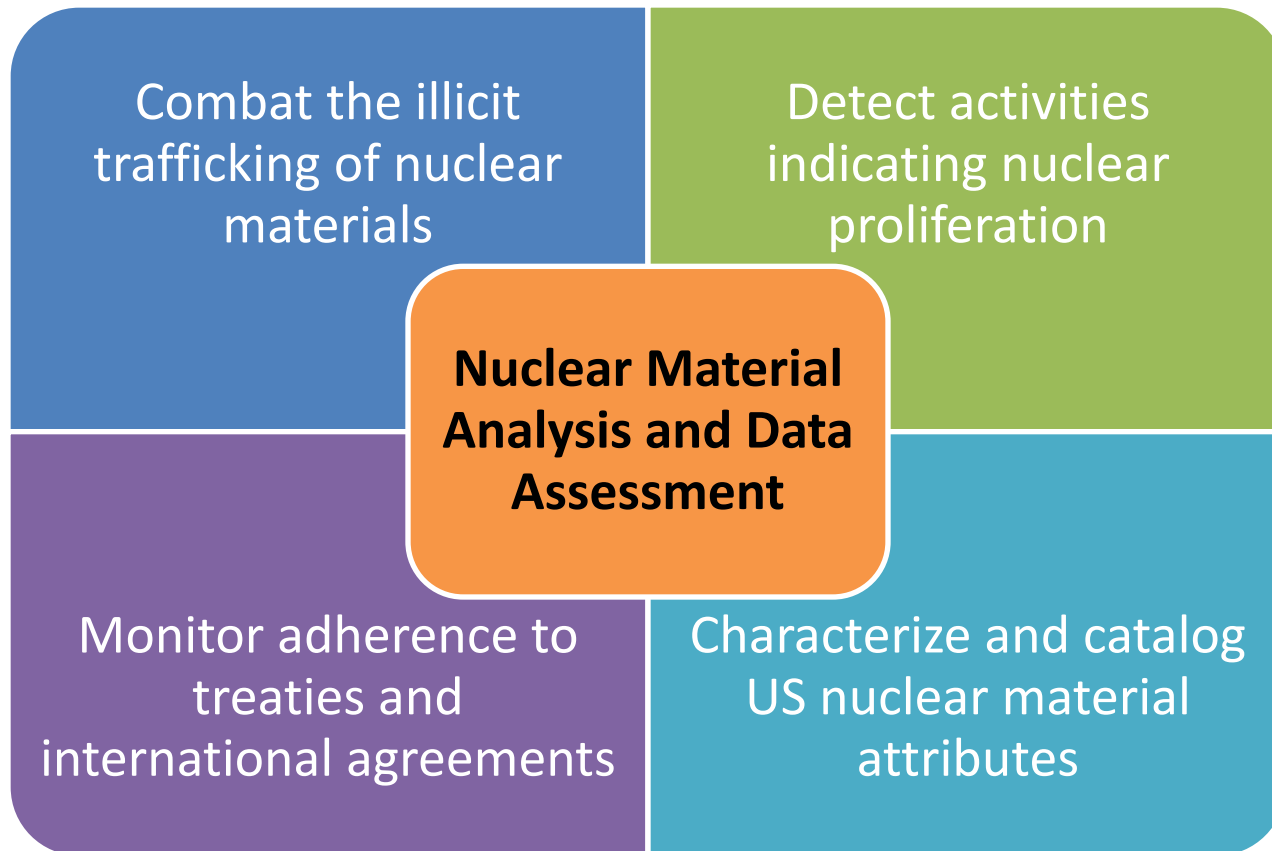


Post-detonation



Device

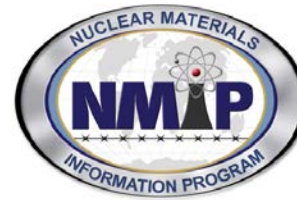
Pre-Detonation Nuclear Forensics Capabilities Support a Wide Range of Activities



Operational Nuclear Forensics Designation

The Bulk Sample Analysis Program (BSAP) is responsible for providing chemical and physical measurements of bulk special nuclear material in support of interagency partners, including the FBI, DOE-IN, DHS, and other customers engaged in nuclear forensics. The BSAP mission supports technical nuclear forensics by maintaining and providing laboratory analysis capabilities commensurate with customer needs and through the sustainment of a dynamic special nuclear material forensics response capability as identified in the requirements of the National Security Presidential Directive-17/Homeland Security Presidential Directive-4 (NSPD-17/HSPD-4), *National Strategy to Combat Weapons of Mass Destruction*.

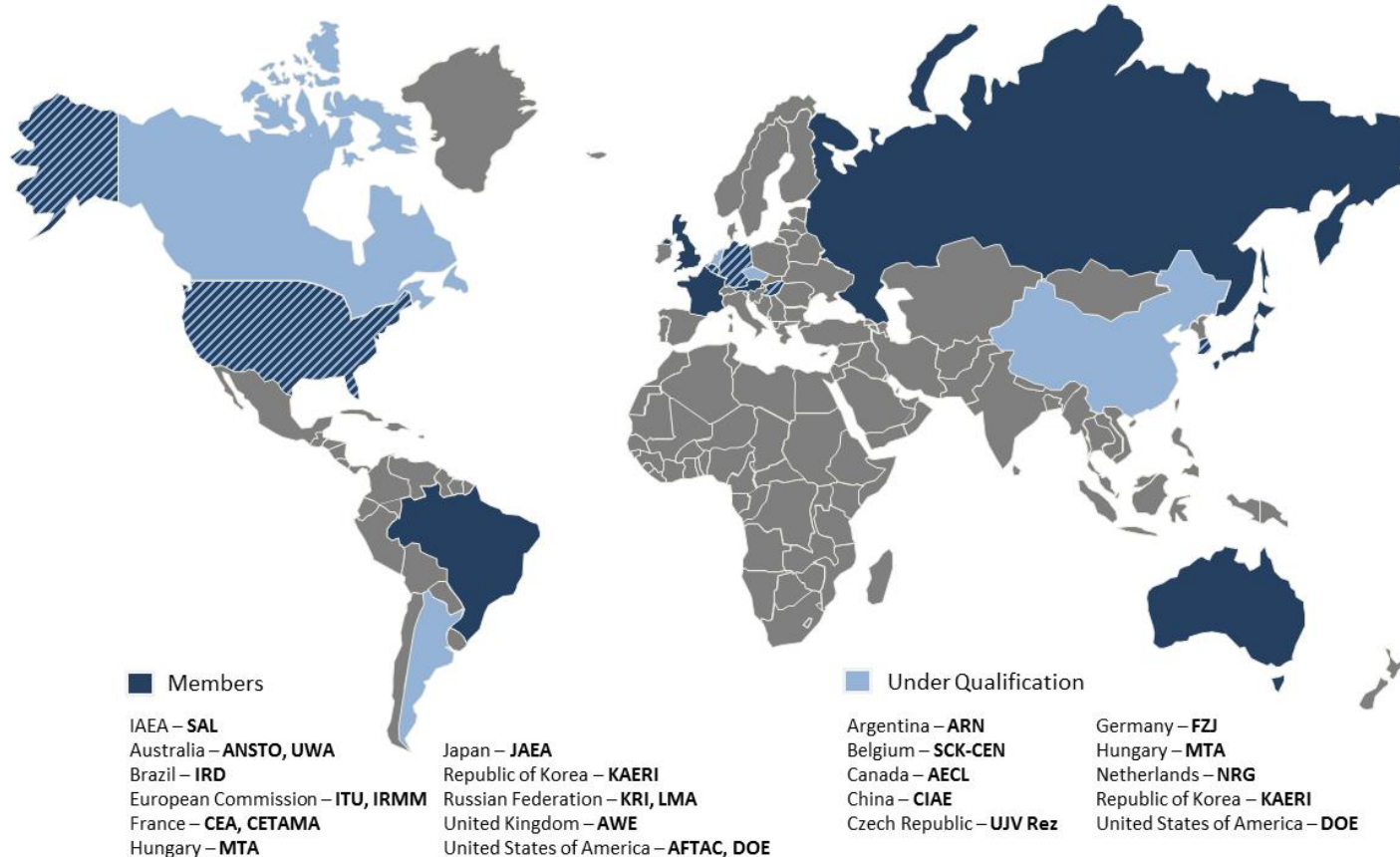
- FBI/CBRNSU
- DOE/NNSA/NA-45
- DOE-IN/NMIP
- DHS/NTNFC & BSAP Partner Laboratories (LANL and LLNL)



Radioanalytical capabilities - application to nonproliferation forensics

IAEA Network of Analytical Laboratories

Updated August 2014



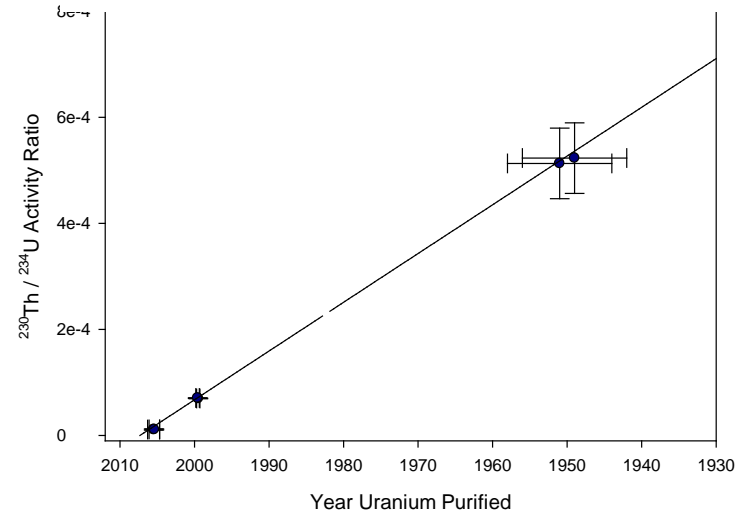
Current LANL International Engagements

- Capability Development – S. Africa
- Nuclear Forensics Training – IAEA Methodologies Course & Kazakhstan engagement
- Document Development – IAEA Technical Documents
- Radiochronometry – CEA, JAEA, CIAE, KAERI, ANSTO, EU
- Nuclear Material Analysis – JAEA
- Morphology – JAEA
- Light Stable Isotopes – Russia

Current Bilateral Engagements– Uranium Age Dating

- Uranium age determination is an essential measurement for nuclear forensics assessments
- No reference materials exist for qualifying age dating methods
- Current radiochronometry engagements – JAEA, CEA, EU, CIAE, KAERI ($^{230}\text{Th}/^{234}\text{U}$ chronometer)
- Proposed radiochronometry engagements - JAEA, CEA, EU, ANSTO ($^{231}\text{Pa}/^{235}\text{U}$ chronometer)
- These DOE/NNSA & partner organization joint measurement exercises help to establish international consistency in uranium age dating measurements

Age of Several Uranium Oxide Samples



Z. Varga, et al., "Validation of Reference Materials for Uranium Radiochronometry in the Frame of Nuclear Forensics" *Applied Radiation and Isotopes*, In Press

A. Gaffney et al., " ^{230}Th - ^{234}U age dating of bulk uranium for nuclear forensics" *Journal of Radioanalytical and Nuclear Chemistry*, Submitted

Several facilities to work with materials of all quantities

All facilities house ongoing missions that exercise analytical capabilities routinely

TA-48



Sigma



CMR

TA-55 & RLUOB



< 10¹¹ atoms Pu-239

10⁹ – 10²² atoms Pu-239

RC-45



RC-1, Sigma



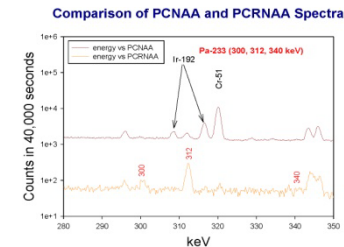
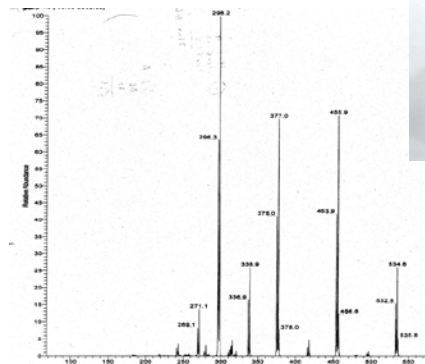
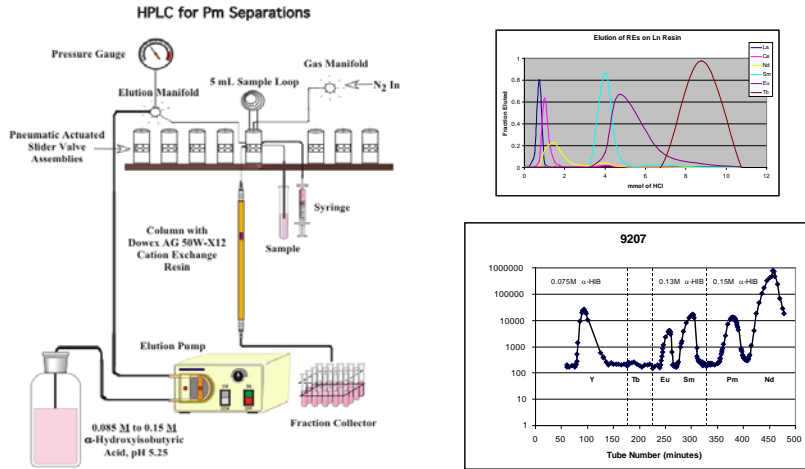
Nuclear facilities (CMR, PF-4)



> 10²² atoms Pu-239

Radiochemistry

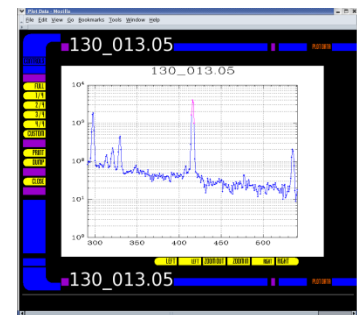
- Single or multi-element
- Elementally separated radiochemistry
- Standardized procedures
- Refractory matrices



Count Room capabilities

Provides qualitative and quantitative assay of gamma, beta, and alpha-emitting radionuclides in a variety of matrices and over a wide range of activity levels,

- Trace levels to $>10^{13}$ fissions
- Specializing in fission product measurements,
- Operates 24x7x365
- Sample receipt and handling protocols
- Makes $\sim 70,000$ measurements annually
 - 30,000 high resolution gamma collections
- 30 non-automated high-resolution gamma-ray spectrometers, some highly specialized
- 10 custom automated high-resolution gamma-ray spectrometers,
- 6 custom automated beta counters.
- 6 custom non-automated beta counters (3 ~ 0.2 CPM Bkg)
- ~ 90 Alpha spectrometers,
- Batch and interactive analysis codes,
- Relational database with web-based visualization tools.



Clean chemistry and mass spectrometry



-State-of-the-art mass spectrometry instrumentation

- TIMS
 - IsotopX Isoprobe -T/Phoenix (7 inst.)
- ICPMS
 - HR-ICPMS - Thermo Element (4 inst.)
 - MC-ICPMS - Thermo Neptune (3 inst.)
 - Nu Inst. Plasma II
- Light Stable Isotopes
 - Thermo MAT 253
 - Thermo Delta V



-10,000 sq. ft. of clean lab space, ideally suited for low-level routine analyses

- Perchloric acid hoods
- Dry and wet ashing

- DOELAP accreditation; full quality envelope

- Many years of routine trace environmental and bioassay monitoring experience

-Expertise in the development and implementation of new procedures

Analysis and characterization of actinide materials

Onsite Analytical Chemistry and Sample Management



Coordinate sample receiving, shipping, and distribution at TA-55 and CMR

Onsite radiochemical and trace analysis

Assay and Classical Chemistry



Coulometric titration
Ceric titration
Pu (III) and Pu (IV)
U Assay by Davies Gray
Fe and Si determination
Loss on Ignition (LOI)
Free acid determination
Standard preparation

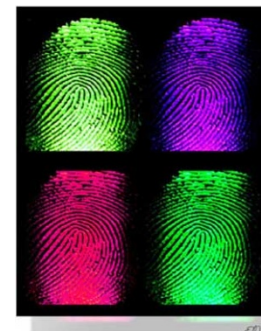
Plasma Spectroscopy

Inductively Coupled Plasma-Mass Spectrometry
Inductively Coupled Plasma- Atomic Emission Spectrometry



DC Arc Emission
Cold-Vapor Atomic Fluorescence

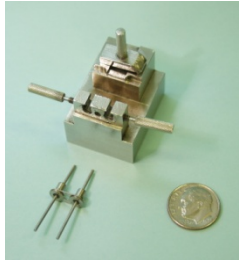
X-Ray Fluorescence (XRF) and X-Ray Diffraction (XRD)



Fingerprint Detection Technology

Analysis and characterization of actinide materials

Mass Spectrometry



High-Precision Gas Mass Spectrometry

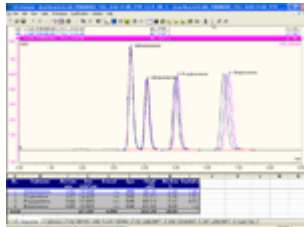


Radiochemistry and Nondestructive Analysis

Alpha and gamma spectrometry
Gross alpha, liquid scintillation



Interstitial Analysis & Ion Chromatography



Fluoride, chloride, nitrite
nitrate, phosphate, sulfate
oxalate and perchlorate

carbon, oxygen, hydrogen
sulfur, moisture, and tritium



Laboratory Information Management System and Quality Assurance

Oracle SQL*LIMS
Sample/nuclear material
tracking and data
management



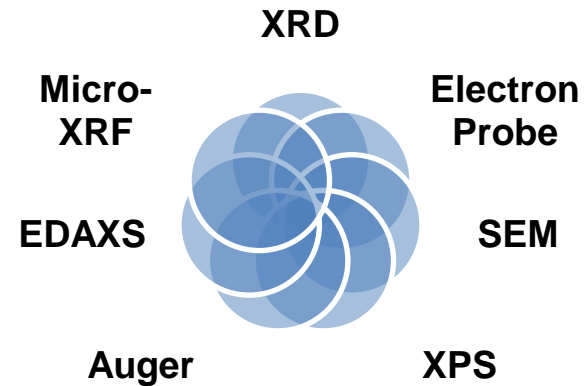
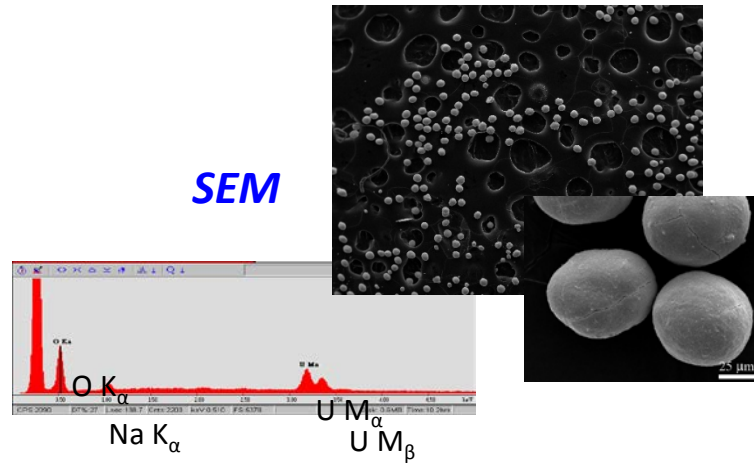
Quality Assurance and Control
Record Management
Document Control
Training



Operated by the Los Alamos National Security, LLC for the DOE/NNSA

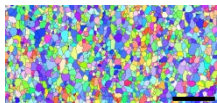


Physical characterization tools for bulk materials

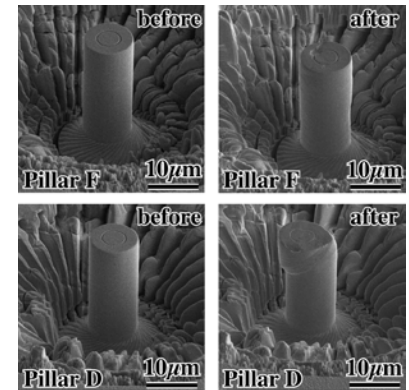
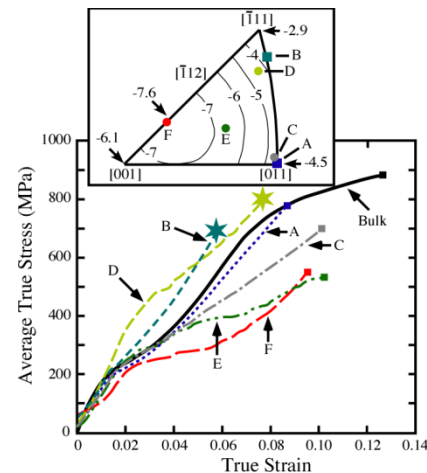
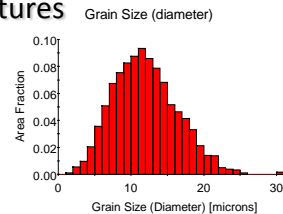


Materials processing techniques leave unique signatures in microstructures:

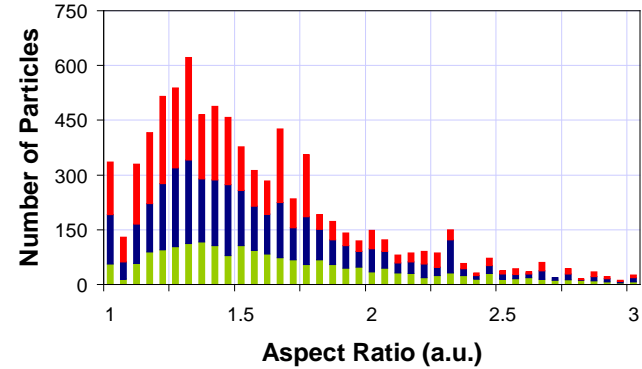
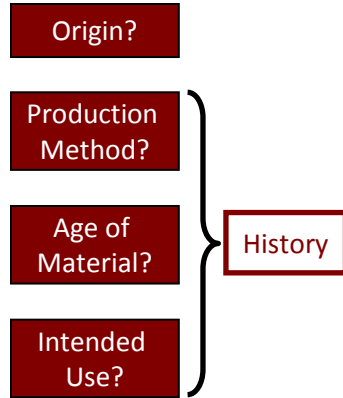
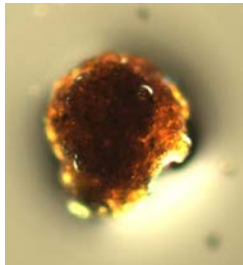
- Grain Size/Morphology
- Inclusion Distribution/Morphology
- Microstructural Texture
- Mechanical Properties
- Surface Features



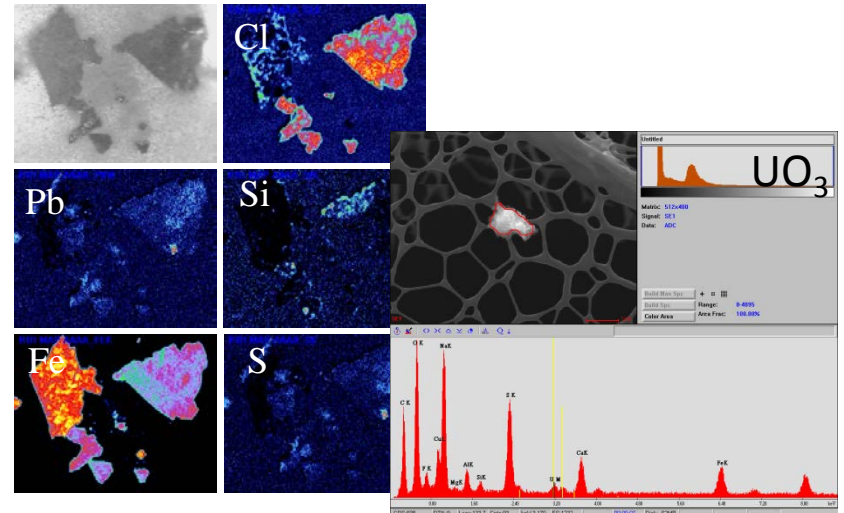
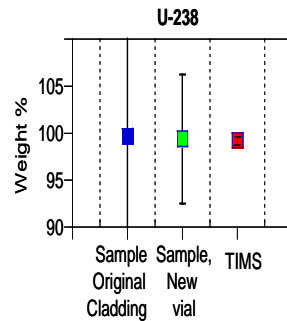
100 μm



Materials analysis – application to forensics

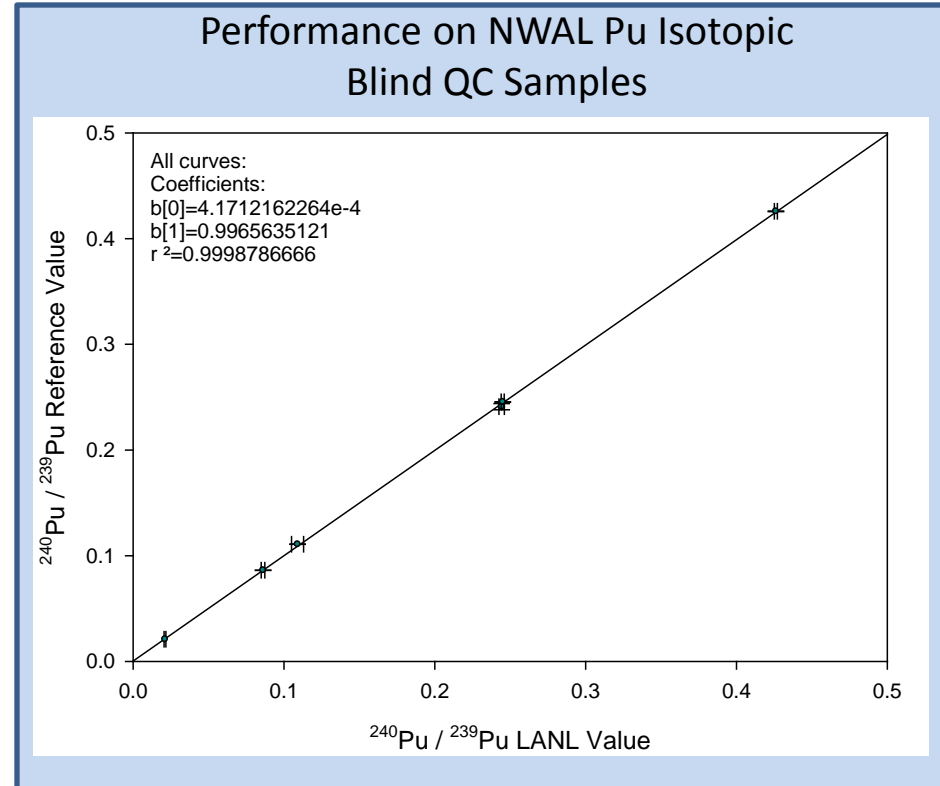


Nuclide	Sample (μg/g)
²³⁴ U	5.5×10^1
²³⁵ U	5.7×10^3
²³⁸ U	7.9×10^5
²³⁸ Pu	7.4×10^{-4}
²³⁹ Pu	2.5×10^1
²⁴⁰ Pu	1.0×10^{-2}
²⁴² Pu	2.5×10^{-4}



Sample management, QA/QC

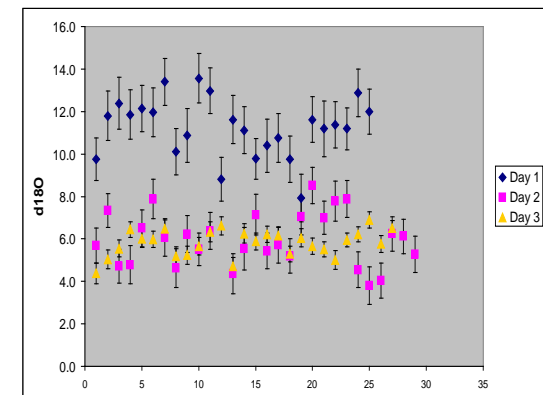
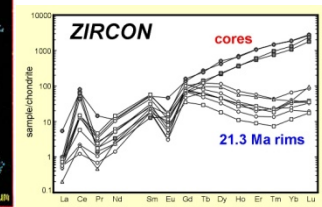
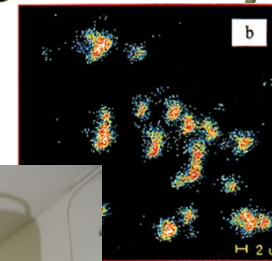
- Bulk analysis and radiochemistry analytical capabilities operate under specific quality programs funded by sponsors.
- Accreditations include: DOELAP, NQA-1, QC-1, and ISO 17025,
- Need for expanded SRM program, laboratory intercomparisons



Investments in modern capabilities

- Neptune MC-ICP-MS
- NU Instruments MC-ICP-MS
- IsotopeX IsoprobeT MC-TIMS
- FEI SEM w/WDS & EDS
- Element XR ICP-MS
- Cameca 1280 multi-collector SIMS
- Fs – laser ablation system

- Dual-Clover high resolution gamma-ray spectrometers



Future technical interests for collaboration

- Advanced analytical methods
- Identification and validation of material signatures from fuel cycle processes
- Methods for archiving nuclear materials analysis data and extracting information from large datasets
- Enhanced QA through the development of new standards and materials exchanges
- Comparative methods for evaluating and assessing nuclear forensics data